THE EFFECT OF RICE HUSKS AS FILLER IN POLYMER MATRIX
COMPOSITES

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ABSTRACT

In this study, rice husks filler polyester composites were produced with rice husks as filler and unsaturated polyester resin as the matrix. Several weight percentages of filler loadings were used; 10 wt%, 15 wt%, 20 wt% and 25 wt% in order to gain insights into the effect of filler content on the mechanical properties and water intake of the composites. The surface fractured of the tensile test specimens were analysed by using Scanning Electron Microscopy. From the findings, the tensile strength of the rice husks filled unsaturated polyester resin composites was found to be decrease as the filler loading increased. However, as it increased to 25 wt%, the strength was slightly increased. As for the young modulus, it showed remarkable increased for 15 wt% of rice husks. Nevertheless, it was decreasing as the rice husks were continually increased up to 25 wt%. Water absorption test was also conducted and the results showed that the composites absorb more water as the weight percentage of rice husks increased, which attributed to the ability of rice husks fillers to absorb water.

ABSTRAK

Dalam kajian ini, sekam padi komposit poliester telah dihasilkan menggunakan sekam padi sebagai pengisi dan resin poliester tidak tepu sebagai matriks. Tahap-tahap jumlah peratusan berat sekam padi diubah kepada 10, 15, 20 dan 25 peratus berat untuk mengetahui kesan jumlah berat pengisi terhadap sifat mekanikal dan penyerapan air komposit. Permukaan retak pada sampel untuk ujian kekuatan tegangan kemudiannya di analisis menggunakan mikroskop pengesan elektron. Daripada keputusan, didapati bahawa kekuatan tegangan sekam padi terisi komposit poliester tidak tepu berkurang apabila berat sekam padi bertambah. Bagaimanapun, ketika ia meningkat kepada 25 peratus berat, kekuatan tegangan komposit menunjukkan sedikit peningkatan. Bagi modulus lenturan, ia menunjukkan peningkatan yang tinggi untuk komposit yang berisi 15 peratus berat sekam padi dan menurun apabila sekam padi terus ditambah sehingga 25 peratus berat. Ujian penyerapan air dijalankan dan menunjukkan bahawa komposit menyerap lebih banyak air apabila peratusan berat sekam padi bertambah. Ini dianggap berpunca daripada keupayaan semulajadi sekam padi untuk menyerap air.

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LIST OF SYMBOLS

wt%	Weight percentage
σ	True stress, local stress
ε	Strain
^oC	degree celcius
A	Cross section area
kN	kiloNewton
l	Length
l_o	Original length
MPa	Megapascal
mm	milimeter
nm	nanometer
P_{break}	Load at break
P_{max}	Maximum load
μm	micrometer

LIST OF ABBREVIATIONS

ASTM	American Society for Testing and Materials
CMC	Ceramic matrix composites
HDPE	High-Density Polyethylene
KBF	Kenaf bast fiber
LLDPE	Linear Low Density Polyethylene
MEKP	Methyl ethane ketone peroxide
MMC	Metal matrix composites
NCO	Cyanate
NMTs	Natural fiber mat thermoplastic composites
OH	Hydroxide
PALF	Pineapple leaf fiber
PET	Polyethylene terephthalate
PLA	Polylactic acid
PLLA	poly-1-lactic acid resin
PMC	Polymer matrix composites
PP	Polypropylene
PU	Polyurethane
RH	Rice husks
RH-PU	Rice husks- polyurethane
RHF–PP	Rice husks flour- Polypropylene
SEM	Scanning electron microscopy
SUV	Sport utility vehicle
UPR	Unsaturated polyester resin

CHAPTER 1

INTRODUCTION

1.1   INTRODUCTION

Composites are materials that comprise strong load carrying material known as reinforcement imbedded in weaker material which known as matrix. Reinforcement provides strength and rigidity, helping to support structural load. The matrix or binder either organic or inorganic maintains the position and orientation of the reinforcement. Significantly, constituents of the composites retain their individual, physical and chemical properties yet together they produce a combination of qualities which individual constituents would be incapable of producing alone.

During the past decades, many polymer composites have been prepared combined with various types of synthetic reinforcing fillers in order to improve the mechanical properties and obtain the characteristics demanded in actual applications (Yao *et al*., 2008). The use of natural filler for the reinforcement of the composites has received increasing attention both by the academic sector and the industry. Natural filler have many significant advantages over synthetic filler and fibres such as their lightweight, low cost, ability to reduce abrasion of machinery and non-toxicity. Currently, many types of natural fillers have been investigated to be used in industry including flax, hemp, wood, wheat, barley, and oats (Bledzki *et al*., 1999). They are now fast evolving as potential alternatives to inorganic or synthetic materials for various applications such as building materials and automotive components (Nourbakhsh *et al*., 2010).

The manufacture of natural fiber composites includes the use of either a thermoset or thermoplastic polymer binder system combined with a natural fiber preform or mat. In automotive applications, the most common system used today is thermoplastic polypropylene, particularly for nonstructural components. Polypropylene is favoured due to its low density, excellent processability, mechanical properties, excellent electrical properties, and good dimensional stability and impact strength (George *et al.*, 2001).

Other than that, rice husks as fillers have advantages over mineral fillers since they are non-abrasive, require less energy for processing and have ability to reduce the density of furnished products. Hence, these composites have attracted much attention, and are becoming increasingly important for the production of a wide variety of cheap lightweight environment friendly composites.

1.2   PROBLEM STATEMENT

Productions of composite often involve huge investment in material acquisition. One way of reducing the production cost but still maintaining the properties of the composite is by using natural filler such as rice husks. Rice husks had been choose due their availability, low cost, low density, high specific strength and modulus, and recyclability (Ismail *et al.*, 2001; Ndazi 2001). In the current work, the effect of rice husk as a filler in polymer matrix composites had been studied.

1.3   OBJECTIVE

These are the objectives of the current work:

(i)   To determine the effect of different percentage of filler in polyester matrix material using rice husks.

(ii)   To evaluate the mechanical and physical properties of polyester matrix material using rice husks.

1.4   SCOPES

The scopes of this project include:

(i)   Preparation of rice husks

(ii)   Preparation of rice husks-unsaturated polyester resin (RH-UPR) composites with different filler loading

(iii)   Several tests such as (a), (b) and (c) had been carried out in order to determine the properties of the composites.

   (a) Tensile Test (ASTM D3039)

   (b) Water Absorption Test (ASTM D570)

   (c) Scanning Electron Microscope (SEM)

CHAPTER 2

LITERATURE REVIEW

2.1   INTRODUCTION

Development of natural material as fillers in the composites has been a subject of interest for the past few years. The use of natural filler can lead to superior materials, in some cases at a premium cost, or they can be a means for achieving improved properties while maintaining low costs. The low cost and the good performance of polyester products have increased the utilization of this polymer in many applications from household materials till the automotive industry. This chapter reviews the topics related that will be used in this research.

2.2   COMPOSITE

Composites are materials that comprise strong load carrying material (known as reinforcement) imbedded in weaker material (known as matrix). Reinforcement provides strength and rigidity, helping to support structural load. The matrix or binder (natural or synthetic) maintains the position and orientation of the reinforcement. Significantly, constituents of the composites retain their individual, physical and chemical properties yet together they produce a combination of qualities which individual constituents would be incapable of producing alone (Hull *et al*., 1996).

Composites are combinations of two materials in which one of the materials, called the reinforcing phase, is in the form of fibers, sheets, or particles, and are embedded in the other materials called the matrix phase. The reinforcing material and the matrix material can be metal, ceramic, or polymer. Typically, reinforcing materials are strong with low densities while the matrix is usually a ductile, or tough, material. If the composite is designed and fabricated correctly, it combines the strength of the reinforcement with the toughness of the matrix to achieve a combination of desirable properties which is not available in any single conventional material (Schaffer *et al.*, 1999).

2.3   CHARACTERISTICS OF THE COMPOSITES

Composites consist of one or more discontinuous phases embedded in a continuous phase. The discontinuous phase is usually harder and stronger than the continuous phase and is called the 'reinforcement' or 'reinforcing material', whereas the continuous phase is known as the ' matrix'.

The shape of the discontinuous phase can be spherical, cylindrical, or rectangular cross-sanctioned prisms or platelets. The size and size distribution which controls the texture of the material and volume fraction determine the interfacial area, which plays an important role in determining the extent of the interaction between the reinforcement and the matrix. While concentration, usually measured as volume or weight fraction, determines the contribution of a single constituent to the overall properties of the composites. The concentration distribution and orientation of the reinforcement will affect the properties such as the strength of the composites. It is not only the single most important parameter influencing the properties of the composites, but also an easily controllable manufacturing variable used to alter its properties (Hinrichsen *et al.*, 2000).

2.4   TYPE OF COMPOSITES

There are three main types of composites which are polymer matrix composites, metal matrix composites and ceramic matrix composites. The composites can be

grouped into categories based on the type of the matrix used. If the matrix used was from polymer group such as polyester, the composite then known as polymer matrix composite. Same goes to the ceramic and metal matrix. Methods of fabrication also vary according to the physical and chemical properties of the matrices and reinforcing fibers (Amar *et al.*, 2005). Each type of the composite has their own advantages and disadvantages which are going to be discussed in section 2.4.1, 2.4.2, and 2.4.3.

2.4.1 Polymer Matrix Composites (PMC)

The most common advanced composites are PMC. These composites consist of a polymer thermoplastic or thermosetting reinforced by fiber or filler. These materials can be fashioned into a variety of shapes and sizes. They provide great strength and stiffness along with resistance to corrosion. The reason for PMC become favourable is their low cost, high strength and simple manufacturing principles.

Besides, as mentioned, polymers consist of thermosets and thermoplastics. Thermosets have qualities such as a well bonded three dimensional molecular structure after curing. They decompose instead of melting or hardening. Merely changing the basic composition of the resin is enough to alter the conditions suitably for curing and determine its other characteristics. They can be retained in a partially cured condition too over prolonged periods of time, rendering thermosets very flexible. Thus, they are most suited as matrix bases for advanced conditions fiber reinforced composites. Thermosets find wide ranging applications in the chopped fiber composites form particularly when a premixed or moulding compound with fibers of specific quality and aspect ratio happens to be starting material as in epoxy, polyester and phenolic polyamide resins.

2.4.2 Metal Matrix Composites (MMC)

MMC as the name implies, have a metal matrix including carbon and silicon carbide. High strength, fracture toughness and stiffness are offered by metal matrices than those offered by their polymer counterparts. They can withstand elevated temperature in corrosive environment than polymer composites. However the guiding

aspect for the choice depends essentially on the matrix material. Most metals and alloys could be used as matrices and they require reinforcement materials which need to be stable over a range of temperature and non-reactive too. Examples of matrices in such composites include aluminum, magnesium and titanium. These metals are mainly reinforced to suit the needs of design. For example, the elastic stiffness and strength of metals can be increased, while large coefficient of thermal expansion and electrical conductivities of metals can be reduced by the addition of fibers such as silicon carbide.

2.4.3 Ceramic Matrix Composites (CMC)

Ceramics can be described as solid materials which exhibit a very strong ionic bonding in general and in few cases covalent bonding. CMC have ceramic matrix such as alumina, calcium, alumina silicate reinforced by silicon carbide. High melting points (above 1600ºC), good corrosion resistance, stabil at elevated temperatures and high flexural strength (550 MPa), render ceramic based matrix materials a favourite for applications requiring a structural material that can withstand at temperatures above 1500ºC. These composites offer the same high temperature (up to 1100°C) tolerance of super alloys but without such a high density (less than 4.5 g/cm^3) (Levitin, 2006). CMC are the obvious choice for high temperature applications. However, other than naturally resistant to high temperature, ceramic materials also have a tendency to become brittle and to fracture. The brittle nature of ceramics makes composite fabrication difficult.

2.5 CONSTITUENTS OF COMPOSITES

2.5.1 Matrices

The role of matrix in a composite is to transfer stresses between the fillers, to provide a barrier against an adverse environment and to protect the surface from mechanical abrasion. The matrix plays a major role in the tensile load carrying capacity of a composite structure. The binding agent or matrix in the composite is of critical importance. Four major types of matrices have been reported which are polymeric, metallic, ceramic and carbon. Most of the composites used in the industry nowadays are

based on polymer matrices. Polymer resins have been divided into two categories which are thermosetting and thermoplastics. Figure 2.1 helps to classify the matrices.
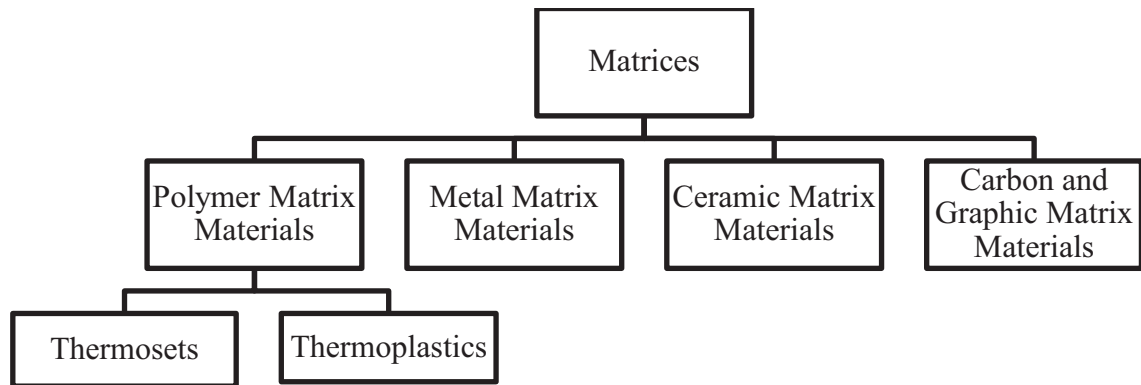
Figure 2.1: Classification of matrices

2.5.2 Reinforcement elements

Reinforcing constituents in composites, as the word indicates, provide the strength to the composite. They also serve certain additional purposes like heat resistance or conduction, resistance to corrosion and provide rigidity. These following three categories are the reinforcement elements that are commonly used:

(i) Fiber Reinforced Composites

A fiber can be characterized by its length. The fibers can be in a form of long continuous fibers, or they can be discontinuous fibers, particles, whiskers and even weaved sheets. The dimensions of the reinforcement determine its capability of contributing its properties to the composite. Fibers are very effective in improving the fracture resistance of the matrix since a reinforcement that having a long dimension will discourages the growth of incipient cracks normal to the reinforcement that might otherwise lead to failure, particularly with brittle matrices. Fiberglass is likely the best know fiber reinforced composite but carbon-epoxy and other advanced composites also fall into this category.

Fibers are usually combined with ductile matrix materials, such as metals and polymers, to make them stiffer. Fibers also added to brittle matrix materials like ceramics to increase the toughness. The lengths to diameter ratio of the fiber, the strength of the bond between the fiber and the matrix, as well as the amount of fiber are variables that affect the mechanical properties. It is important to have a high length to diameter aspect ratio so that the applied load is effectively transferred form the matrix to the fiber.

(ii)   *Particle Reinforced Composites*

As the name itself indicates, the reinforcement is of particle nature (platelets are also included in this group). It may be spherical, cubic, tetragonal, a platelet, or of other regular or irregular shape, but it is approximately same. In general, particles are not very effective in improving fracture resistance but they enhance the stiffness of the composite to a limited extent. Particle fillers are widely used to improve the properties of matrix materials such as to modify the thermal and electrical conductivities, improve performance at elevated temperatures, reduce friction, increase wear and abrasion resistance, improve machinability, increase surface hardness and reduce shrinkage.

The particle size can be found from 0.1 µm to 30 nm size depends on the particle itself. Zhang *et al*. (2006) found that for the Si nano-particles, they have an average size of 25 nm while Makame *et al*. (2005) reported on using average particle sizes of cashew nut ranging from 0.1 µm to 0.4 µm. In this case, the particles carry a major portion of the load. The particles are used to increase the modulus and decrease the ductility of the matrix. An example of particle reinforced composites is an automobile tire which has carbon black particles in a matrix of polyisobutylene elastomeric polymer.

Particle reinforced composites are much easier and less costly than making fiber reinforced composites. With polymeric matrices, the particles are simply added to the polymer and melt in an extruder or injection molder during polymer processing. The reinforcing particles are added to a molten metal before it is cast.

(iii) **Dispersion Strengthen Composites**

In dispersion strengthened composites, small particles length on the order of 10.5 mm to 12.5 mm (length) and 10.4 mm (diameter) are added to the matrix material. These particles act to help the matrix resist deformation. This makes the material harder and stronger. Consider a metal matrix composite with a fine distribution of very hard and small secondary particles. The matrix material is carrying most of the load and deformation is accomplished by slip and dislocation movement. The secondary particles impede slip and dislocation and, thereby, strengthen the material. The mechanism is that same as precipitation hardening but effect is not quite as strong. However, particles like oxides do not react with the matrix or go into the solutions that have high temperatures (1200°C) so the strengthening action is retained at elevated temperatures (Sinha, 2000).

2.6   POLYMERS

The word polymer literally means many parts. A polymeric solid material may be considered to be one that contains many chemically bonded parts or units that being bonded together to form a solid. The term polymer refers as long chain molecule that is composed of a large number of repeating units of identical structure (Smith *et al*., 2006). This covalent bonding is formed via polymerization process and can have shape of linear or even more complicated networking. There are two industrially important plastic polymer materials which are plastics and elastomers. Plastics are large and varied group of synthetic materials that are processed by forming or molding into shape. Plastic also can be divided into two classes; thermoplastics and thermosetting plastics which are going to be discussed in section 2.6.1 and 2.6.2.

2.6.1   Thermoplastics

Thermoplastics are polymers that require heat to make them processable. They become soft and deformable upon heating. After cooling, such materials retain their shape. In addition, these polymers may be reheated and reformed, often without significant changes in their properties (Sinha, 2000). The high temperature plasticity is

due to the ability of the molecules to slide past one another, which is an example of a thermally activated. In this sense, thermoplastics materials are similar to metal that gain ductility at high temperature like creep deformation. Thermoplastics also retain good strength and stiffness. Some examples of thermoplastics polymer are polyethylene, polystyrene, polyamides, nylons and polypropylene.

2.6.2   Thermosetting

Thermoset is a hard and stiff cross linked material that does not soften or become moldable when heated. Thermosets are stiff and do not stretch the way that elastomers and thermoplastics do. Several types of polymers have been used as matrices for natural fiber composites (Sinha, 2000). Most commonly used thermoset polymers are epoxy resins and other resins which is unsaturated polyester resins as in fiberglass, vinyl ester, phenolic epoxy, novolac and polyamide (Chawla *et al.*, 1997).

Prior to cure, thermosets were liquid or made to flow under pressure to any desired form (Matthews *et al.*, 1999). Once cured, a tightly bound three dimensional network structure was formed in the resin and hence the resin cannot be melted, reshaped and reprocess by heating (Hull *et al.*, 1996). Therefore, during composite manufacturing, the impregnation process followed by the shaping and solidification were done before the resin begin to cure (Courtney, 1990). Epoxy resin, phenolic polyamide resins and unsaturated polyester resin are common examples of thermosets polymer.

2.7   UNSATURATED POLYESTER RESIN (UPR)

UPR has reactive double carbon-carbon covalent bonds that can be cross linked to form thermosetting materials. The linear unsaturated polyesters are usually cross-linked with vinyl type molecules such as styrene in the presence of free radical curing agent. Peroxide curing agents are mostly used, with methyl ethyl ketone peroxide (MEKP) usually being used for the room temperature curing of polyester. In addition, The UPR is low viscosity materials that can be mixed with high amounts of fillers and

reinforcements. For example, unsaturated polyester may contain as high as about 80 wt% of glass fibre reinforcement.

The applications of polyester based material are extensive. This material used for making automobile panels and body parts and also used for small boat hulls (Dejan *et al*., 2009). Unsaturated reinforced polyesters are also used for pipes, tanks, and ducts where good corrosion resistance required. In general, polyesters are known for the good adhesion, chemical and heat resistance, good mechanical properties and very good electrical insulating properties (Daimler *et al*., 2000). Other than that, the reinforcement of polyesters with cellulosic fibers has been widely reported. Some of them were polyester-pineapple leaf by George *et al.* (2000) and polyester-jute by Joseph (1999) and Roe *et al.* (1985). The properties of typical thermoset polymers for natural composites are listed in Table 2.1. It contains the basic properties of the thermoset.

Table 2.1: Properties of typical thermoset polymers for natural fiber composites

Property	Polyester resin	Vinylester resin	Epoxy
Density (g/cc)	1.2-1.5	1.2-1.4	1.1-1.4
Elastic Modulus (Gpa)	2-4.5	3.1-3.8	3-6
Tensile Strength (Mpa)	40-90	69-83	35-100
Compressive Strength (Mpa)	90-250	100	100-200
Elongation (%)	2	4-7	1-6
Cure Shrinkage (%)	4-8	-	1-2
Water Absorption (24 hours at 20°C)	0.1-0.3	0.1	0.1-0.4
Izod Impact, Notched (J/cm)	0.15-3.2	2.5	0.3

Source: Daimler *et al*. (2000)

2.8   NATURAL FILLER REINFORCED

The use of natural filler for the reinforcement of the composites has received increasing attention both by the academic sector and the industry. Natural filler have many significant advantages over synthetic fillers such as low cost, low density, biodegradability, non-abrasiveness and recyclability (Rana *et al*., 2003). Due to these

advantages, the utilization of natural fillers derived from agricultural sources such as pineapple, jute, rubber wood, oil palm fruit bunch and rice husk ash in polymer composites has become rapidly expanded. Natural fillers also provide positive environmental benefits with respect to raw material utilization where large amount pollution causing by of agricultural wastes can be reduce. The typical properties of filler are listed in Table 2.2.

Table 2.2: Typical Filler Properties

Category	Description
Filler Shape	Spherical, cubical, irregular, block, plate, flake, fibre, mixtures of different shapes.
Distribution	Monodisperse, designed mixture of sizes, Gaussian distribution, irregular distribution.
Chemical	May be inorganic and of an established chemical composition
Compositions	May also be a single element, natural products or mixtures of different materials in unknown proportions (waste and recycled materials).
Physical State	All materials discussed are solids but they might be available in a pre-dispersed state.
Filler Size	Range from a few nanometers to tens of millimetres

Source: George (1999)

Generally, plant or vegetable fibers are used to reinforce polymer matrices. Plant fibers are a renewable resource and have the ability to be recycled. The classifications of fibers are given in Figure 2.2.